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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<p>(21) International Application Number: <b>PCT/US91/09067</b></p> <p>(22) International Filing Date: <b>10 December 1991 (10.12.91)</b></p> <p>(30) Priority data: 626,491 12 December 1990 (12.12.90) US 762,068 23 September 1991 (23.09.91) US</p> <p>(71) Applicant: E.I. DU PONT DE NEMOURS AND COMPANY [US/US]; 1007 Market Street, Wilmington, DE 19898 (US).</p> <p>(72) Inventor: TANNENBAUM, Harvey, Paul ; 9 Overbrook Parkway, Wynnewood, PA 19096 (US).</p> <p>(74) Agents: BURGESS, Richard, H. et al.; E.I. du Pont de Nemours and Company, Legal/Patent Records Center, 1007 Market Street, Wilmington, DE 19898 (US).</p>		<p>(81) Designated States: AT (European patent), AU, BE (European patent), BR, CA, CH (European patent), DE (European patent), DK (European patent), ES (European patent), FR (European patent), GB (European patent), GR (European patent), IT (European patent), JP, KR, LU (European patent), MC (European patent), NL (European patent), SE (European patent), SU<sup>+</sup>.</p> <p><b>Published</b> <i>With international search report. With amended claims.</i></p>	
<p>(54) Title: <b>NON-STICK COATING SYSTEM WITH PTFE AND PFA OR FEP FOR CONCENTRATION GRADIENT</b></p> <p>(57) Abstract</p> <p>Improved non-stick coating systems can be applied to untreated smooth substrate with a primer of a polytetrafluoroethylene having a melt viscosity over <math>10^{10}</math>Pa Sec plus a copolymer of tetrafluoroethylene with hexafluoropropylene or perfluoro alkyl vinyl ether with the melt viscosity of the polytetrafluoroethylene being at least <math>10^2</math>Pa Sec higher than that of the copolymer to give a concentration gradient.</p>			

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TITLE

NON-STICK COATING SYSTEM  
WITH PTFE AND PFA OR FEP FOR CONCENTRATION GRADIENT  
BACKGROUND OF THE INVENTION

This invention relates to coatings systems, such as for cookware, which minimize sticking. More particularly, it relates to multilayer 5 coating systems that can be used directly on smooth, untreated substrates and which provide concentration gradients within the coating.

Generally in the art a metal or glass substrate is roughened by some means before the first layer of coating is applied so that mechanical bonding will assist chemical adhesive means in holding the coating onto the 10 substrate. Typical roughening means include acid etching, sand-blasting, grit-blasting, and baking a rough layer of glass, ceramic or enamel frit onto the substrate. The problem of adhesion of non-stick coatings to substrates is exacerbated by the nature of the coatings. If the coating is optimized for release to prevent food particles from sticking to it, for easy clean-up after 15 cooking or durability, or to facilitate low friction sliding contact, almost by definition there will be difficulties in making it adhere well to the substrate.

The substrate can be metal, often aluminum or stainless steel used for cookware or industrial applications. It can be glass or ceramic. It might even be plastic for microwave oven cookware, or it could be an 20 industrial article such as a saw made of carbon steel. Whatever the substrate or the application, if it is necessary to roughen the substrate to make the coating adhere, that at least adds cost and can cause other difficulties including creating a rough profile which can protrude or telegraph through the coating. This is especially undesirable when smoothness is sought, such 25 as for saws, steam irons and copier rolls. The environmental cost of disposing of etchant materials can be significant. Sometimes, especially for glass and ceramic substrates, it also can cause unacceptable weakness or brittleness of the substrate.

Means of enhancing adhesion of non-stick coatings to a 30 substrate are illustrated by the following patents.

U.S. 4,049,863 - Vassiliou (1977) teaches a primer containing fluoropolymer, such as polytetrafluoroethylene (PTFE), colloidal silica and a polyamide imide (PAI), along with other constituents, applied by various

techniques to a substrate that is preferably pretreated by grit blasting, flame spraying of metals or metal oxides or frit coating, or to phosphated and chromated metals. The PTFE:PAI ratio can be 1:9. The primer coat is ordinarily applied to a dry film thickness (DFT) of about 2-15 microns ( $\mu\text{m}$ ). After air drying, the primer is topcoated with a conventional fluoropolymer enamel and baked. (Parts, percentages and proportions herein are by weight except where indicated otherwise.)

U.S. 4,087,394 - Concannon (1987) discloses aqueous concentration gradient coatings of fluoropolymer which is 20-80% of a homopolymer or a copolymer of fluorinated ethylene-propylene (FEP) made of 5-100% tetrafluoroethylene (TFE) with 95-0% hexafluoropropylene (HFP), with 80-20% of a film forming polymer which can be PAI. The coating is applied by spraying onto aluminum sheet, or a variety of substrates. Other application techniques are mentioned. Nothing is said about substrate preparation. Although PTFE and FEP are treated as a continuum, there are no suggestions to use a blend such as 50% PTFE, 50% FEP.

15 U.S. 3,928,675 and 3,857,852, both to Tieszen, teach the use of high viscosity ( $>10^{10}$ ) and low viscosity ( $10^4$  poise) ( $10^9$  and  $10^3$  Pa Sec) PTFE along with polyarylene sulfide such as polyphenylene sulfide (PPS) in coatings.

#### SUMMARY OF THE INVENTION

20 The present invention, in certain of its embodiments, provides a coating system comprising a substrate with a multi-layer non-stick coating, comprising a primer, a topcoat, and up to one or more intermediate coats between the primer and the topcoat, wherein:

the substrate is free of contaminants that would prevent  
25 adhesion of the coating,

the primer is applied to the substrate in the form of an aqueous dispersion comprising perfluorocarbon resin and at least one of polyamide imide, and polyether sulfone resins wherein the perfluorocarbon resin comprises 50-90% by weight of a first resin of polytetrafluoroethylene having 30 a melt viscosity of at least about  $10^{10}$  poises plus 50-10% of a second resin of perfluorinated copolymer of perfluoro alkyl vinyl ether, preferably perfluoro propyl vinyl ether, and tetrafluoroethylene (PFA) having a melt viscosity in the range of  $10^3$  to  $10^5$  poises, and

the topcoat and any intermediate coats comprise perfluorocarbon resin.

#### DETAILED DESCRIPTION

The present invention permits not only lower cost by avoiding the roughening of the substrate but also smoother coated surfaces which can be advantageous for release on cookware, and for the gliding effect on steam 5 iron sole plates. Also it can allow elimination of costly polishing of coated copier roll surfaces and application of dispersion PTFE coatings by coil coating and roller coating techniques.

Various embodiments of the invention involve using at least two PTFE resins having different melt viscosities in a primer or a topcoat.

10 One pair of resins has relatively high and low melt viscosity resins. Another has relatively low and lower still melt viscosity resin.

The adhesion of high melt viscosity fluoropolymer coatings to all types of metal substrates, particularly to smooth metal, can be significantly improved through chemically induced stratification or formation of a 15 concentration gradient in the primer.

Addition of perfluorocarbon polymer having a low melt viscosity (MV) in the range of  $10^3$ - $10^8$  poise ( $10^2$  -  $10^7$  Pa Sec), to a primer system composed of PTFE with a high MV of  $10^{11}$  poise ( $10^{10}$  Pa Sec) and a polymeric binder such as polyamide-imide or polyphenylene sulfide, 20 imparts a synergistic effect in which the fluoropolymer stratifies away from the substrate interface allowing the polymeric binder to obtain a higher concentration and degree of cure at the substrate interface resulting in improved adhesion. The required cure temperature to achieve this stratification can be modified by the choice of fluoropolymer.

25 Melt viscosity of perfluoropolymers can be determined by known technique such as that in U.S. Patent 4,636,549 - Gangal et al (1987). See Col. 4, lines 25 - 63.

With use of the coatings of the invention on smooth substrates, treated only by washing to remove grease and any other 30 contaminants which might interfere with adhesion, coating systems of the invention give good food release and good resistance to usual durability tests such as the "tiger paw" abuse cooking tests involving a weighted holder with multiple ball point pen shafts rotating around the inside of a frying pan

during cooking tests. The tests are generally described in U.S. patent 4,252,859, -- Concannon and Vary (1981) col. 2, lines 14-24.

Typical prior art preparation of surfaces to enhance adhesion of a release coating has involved etching or sand or grit blasting to develop a surface profile. The profile is measured in average microinches using a model RT 60 surface roughness tester made by Alpa Co. of Milan, Italy. The 5 profile on typical rolled aluminum after washing to remove grease and contaminants is 16-24 microinches (.6 - 0.96  $\mu\text{m}$ ). The profile on steel varies more widely but is typically less than 50 microinches (2  $\mu\text{m}$ ). On both steel and aluminum, before a release coating is applied the profile typically is increased to over 100 micro inches (4  $\mu\text{m}$ ), preferably for aluminum for some 10 uses to 180-220 micro inches (7.2 - 8.8  $\mu\text{m}$ ). Thus, the present invention is particularly useful with steel or aluminum substrates having a profile of less than 100, preferably less than 50 micro inches (less than 4  $\mu\text{m}$ , preferably less than 2  $\mu\text{m}$ ).

Similar effects can be achieved using a low MV (at least  $10^6$  15  $10^5$  Pa Sec) PTFE with a lower still MV ( $10^3$  to  $10^5$  poise or  $10^2$  to  $10^4$  M Pa Sec) PTFE. To obtain stratification, it is desirable to have a difference of at least  $10^2$  poise in melt viscosities of the two PTFE's.

The primers of the invention can also be used on substrates roughened in various ways known in the art to make coating systems even 20 better than without such undercoats. This can combine improved chemical adhesion with mechanical effects to produce products that may be superior.

In the following examples, the polyamide imide, colloidal silica and dispersions are known in the art and preferably are those of U.S. Patent 4,049,863 - Vassiliou (1977); the PFA is that generally disclosed in U.S. 25 Patent 4,253,859--Concannon and Vary (1981), but with a melt viscosity in the ranges of  $2-4 \times 10^4$  poises, preferably in the form of a pulverized powder or a dried dispersion, either having an average particle size in the range of 20-25  $\mu\text{m}$ ; and the ultramarine blue is that of U.S. Patent 4,425,448 - Concannon and Rummel (1984).

30 The following examples and test data demonstrate this improved adhesion when used as a primer for fluoropolymer topcoats. The fluoropolymers are provided as 60% dispersions in water. As usual, the solids content of dispersions is indicated in the tables. The compositions

were blended by techniques normal in the art and then applied to a smooth, degreased aluminum substrate by spraying.

EXAMPLE 1: FEP/PTFE - Multiple Coat System

Table 1

Composition: 40% FEP/60% Primer PTFE

	<u>Weight</u>	<u>Percent</u>	
5	0.007		Zinc oxide
	0.050		"Afflair 153" titania coated
			mica from EM Industries
10	6.497		Ultramarine Blue pigment
	6.750		"T-30" PTFE from Du Pont
	0.972		"Ludox AM" colloidal silica from
			Du Pont
	4.153		"TE9075" FEP from Du Pont
15	4.641		AI-10 polyamide imide resin from
			Amoco
	67.628		Deionized water
	0.630		"Triton X-100" octyl phenol polyether
			alcohol non-ionic surfactant from
20			Rohm and Haas
	0.655		Diethylethanolamine
	1.309		Triethylamine
	<u>3.614</u>		Furfuryl alcohol
	100.00	<b>TOTAL</b>	

Table 2  
Topcoat

	<u>Weight</u>	
	<u>Percent</u>	
	0.790	"Afflair 153"
	0.389	Channel black pigment
5	0.172	Ultramarine blue pigment
	0.195	Aluminum silicate
	40.704	"T-30" PTFE
	0.442	Cerium octoate
	0.054	Sodium polynaphthalene sulfonate
10	1.834	Diethylene glycol monobutylether
	0.928	Oleic acid
	33.772	Deionized water
	3.480	Triethanol amine
	2.246	Hydrocarbon solvent
15	2.914	"Triton X-100"
	<u>12.080</u>	Acrylic latex of 39 parts by weight
	100.00	terpolymer of methylmethacrylate/57 part ethyl acrylate/4 parts methacrylic acid, dispersion at 40% solids in water, 0.2 sm average particle size
20		

**Application:**

This system is comprised of a primer of PTFE, FEP and 25 polyamide imide which is applied at 5-10 sm dry film thickness (DFT) to a metal surface which has been washed to remove oil and dirt contamination, air dried, and topcoated with a single (15-17.5 sm DFT) or multiple topcoats in thicknesses 12.5-17.5 sm DFT each and having compositions similar to those shown in Table 2. The films are baked 10 minutes at 150° C followed 30 by a high temperature bake for a minimum of 3 minutes over 415° C.

**Testing:**

After application of a single layer coating on smooth, degreased 12 gauge aluminum substrate, cured under varying conditions, the

coated substrate was soaked in boiling water for 20 minutes. The coating is cut down to the substrate, then a person attempts to pull back the coating with his fingernail. In the following Table, P indicates that the coating did not come loose, F indicates that it pulled back at least 1 cm.

Table 3

## Fingernail Adhesion on Smooth Aluminum

Cure (Temp °C/Time - min)

5	780/3	429/5	432/10
	P	P	P

Tests without the FEP led to failure of this coating.

Two different proportions of FEP and PTFE were used as a primer with a topcoat on smooth aluminum cookware which was subjected to tiger paw testing, described above. The number of standard cooking cycles to a rating of 5, determined by coating deterioration, was recorded and presented below along with the percentages of the comparable value for a commercial coating on a grit-blasted substrate run as a control. The results are better than many good commercial products.

Table 4

## Cooking Performance of FEP/PTFE Primer

Cooks to Rating of 5

					% of
	<u>System</u>	<u>DFT</u>	<u>Range</u>	<u>Average</u>	<u>Commercial</u>
20	40% FEP/60% PTFE	1.0-1.1	80-120	95	114
	30% FEP/70% PTFE	1.0-1.1	80-120	103	124

EXAMPLE 2: FEP/PTFE - Multiple Coat SystemTable 1

Composition: 40% FEP/60% PTFE Primer

	<u>Weight</u>	
	<u>Percent</u>	
5	0.007	Zinc Oxide
	0.050	"Afflair 153" titania coated mica
10	6.497	from EM Industries
	6.750	Ultramarine Blue pigment
	0.972	"T-30" PTFE from Du Pont
	4.153	"Ludox AM" colloidal silica
	4.641	from Du Pont
15	67.628	"TE9075" FEP from Du Pont
	0.630	AI-10 polyamide imide resin
	0.655	from Amoco
20	1.309	Deionized water
	3.614	"Triton X-100" octyl phenol polyether
	100.00	alcohol non-ionic surfactant from
25	TOTAL	Rohm and Haas
		Diethylethanolamine
		Triethylamine
		Furfuryl alcohol

CLAIMS

1. A coated substrate comprising a substrate with a multi-layer non-stick coating, comprising a primer, a topcoat, and up to one or more intermediate coats between the primer and the topcoat, wherein;

the substrate is free of contaminants that would prevent adhesion of the coating,

5 the primer is applied to the substrate in the form of an aqueous dispersion comprising perfluorocarbon resin and at least one of polyamide imide and polyether sulfone resins wherein the perfluorocarbon resin comprises 50-90% by weight of a first resin of polytetrafluoroethylene having a melt viscosity of at least about  $10^{10}$  poises plus 50-60% of a second resin

10 selected from perfluorinated copolymer of hexafluoropropylene and tetrafluoroethylene having a melt viscosity in the range of  $10^3$  to  $10^8$  poises ( $10^2$  to  $10^7$  Pa Sec) and perfluorinated copolymer of perfluoro alkyl vinyl ether and tetrafluoroethylene having a melt viscosity in the range of  $10^3$  to  $10^5$  poises, and

15 the topcoat and any intermediate coats comprise perfluorocarbon resin.

2. The coated substrate of claim 1 wherein the melt viscosity of said first resin is at least  $10^{11}$  poises and the melt viscosity of said second resin is in the range of  $10^4$ - $10^5$  poises.

20 3. The coated substrate of claim 2 wherein selected copolymer is a copolymer of hexafluoropropylene and tetrafluoroethylene.

4. The coated substrate of claim 1 wherein the selected copolymer is a copolymer of hexafluoropropylene and tetrafluoroethylene.

5. The coated substrate of claim 1 wherein the substrate is 25 metal selected from aluminum, stainless steel and carbon steel.

6. The coated substrate of claim 5 wherein the substrate before coating has a surface roughness profile less than 2.5 microns.

7. The coated substrate of claim 5 wherein the substrate before coating has a surface roughness profile less than 1.25 microns.

30 8. The coated substrate of claim 1 wherein the primer contains 3-5% colloidal silica, 1-4% surfactant, 15-30% polyamide imide, and 25-55% perfluoropolymer consisting of 60-85% polytetrafluoroethylene, with balance of the perfluoropolymer being the copolymer.

10

9. The coated substrate of claim 1 wherein, before application of the undercoat, the surface of the substrate has been treated to remove contaminants that would interfere with adhesion but has not been etched or mechanically roughened.

10. The coated substrate of claim 1 wherein the primer coating resulting from said aqueous dispersion is not uniform in composition throughout its thickness but has a lower concentration of polytetrafluoroethylene at the interface with the substrate than at the opposite interface.

11. A process of making the coating substrate of claim 1 wherein the coatings are applied to the substrate without completely drying one coating before applying the next, and then the entire coating is cured by heating at at least 350°C.

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## AMENDED CLAIMS

[received by the International Bureau on 21 May 1992(21.05.92);  
original claim 1 amended;  
remaining claims unchanged (1 page)]

1. A coated substrate comprising a substrate with a multi-layer non-stick coating, comprising a primer, a topcoat, and up to one or more intermediate coats between the primer and the topcoat, wherein;  
the substrate is free of contaminants that would prevent  
5 adhesion of the coating,

the primer is applied to the substrate in the form of an aqueous dispersion comprising perfluorocarbon resin and at least one of polyamide imide and polyether sulfone resins wherein the perfluorocarbon resin comprises 50-90% by weight of a first resin of polytetrafluoroethylene having  
10 a melt viscosity of at least about  $10^{10}$  poises plus 50-10% of a second resin selected from perfluorinated copolymer of hexafluoropropylene and tetrafluoroethylene having a melt viscosity in the range of  $10^3$  to  $10^8$  poises ( $10^2$  to  $10^7$  Pa Sec) and perfluorinated copolymer of perfluoro alkyl vinyl ether and tetrafluoroethylene having a melt viscosity in the range of  $10^3$  to  
15  $10^5$  poises, and

the topcoat and any intermediate coats comprise perfluorocarbon resin.

2. The coated substrate of claim 1 wherein the melt viscosity of said first resin is at least  $10^{11}$  poises and the melt viscosity of said second resin is in the range of  $10^4$ - $10^5$  poises.

3. The coated substrate of claim 2 wherein selected copolymer is a copolymer of hexafluoropropylene and tetrafluoroethylene.

4. The coated substrate of claim 1 wherein the selected copolymer is a copolymer of hexafluoropropylene and tetrafluoroethylene.

- 25 5. The coated substrate of claim 1 wherein the substrate is metal selected from aluminum, stainless steel and carbon steel.

6. The coated substrate of claim 5 wherein the substrate before coating has a surface roughness profile less than 2.5 microns.

- 30 7. The coated substrate of claim 5 wherein the substrate before coating has a surface roughness profile less than 1.25 microns.

8. The coated substrate of claim 1 wherein the primer contains 3-5% colloidal silica, 1-4% surfactant, 15-30% polyamide imide, and 25-55% perfluoropolymer consisting of 60-85% polytetrafluoroethylene, with balance of the perfluoropolymer being the copolymer.

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 91/09067

**I. CLASSIFICATION OF SUBJECT MATTER** (If several classification symbols apply, indicate all)<sup>6</sup>

According to International Patent Classification (IPC) or to both National Classification and IPC

Int.Cl. 5 B05D5/08; C09D127/18; C09D179/08; C09D181/06

**II. FIELDS SEARCHED**

Minimum Documentation Searched<sup>7</sup>

Classification System	Classification Symbols	
Int.Cl. 5	B05D ;	C09D

Documentation Searched other than Minimum Documentation  
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**III. DOCUMENTS CONSIDERED TO BE RELEVANT<sup>9</sup>**

Category <sup>10</sup>	Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>
A	EP,A,0 389 966 (SUMITOMO ELECTRIC IND. LTD) 3 October 1990 see claim 1; examples 1-3 ----	1
A	EP,A,0 100 889 (HOECHST AG) 22 February 1984 see examples 4-6 ----	1
A	EP,A,0 056 280 (DU PONT DE NEMOURS) 21 July 1982 see page 8, line 10 - page 10, line 28; claims 1-3 ----	1

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**IV. CERTIFICATION**

Date of the Actual Completion of the International Search

1

14 APRIL 1992

Date of Mailing of this International Search Report

21.04.92

International Searching Authority

EUROPEAN PATENT OFFICE

Signature of Authorized Officer

D. Schüller



**ANNEX TO THE INTERNATIONAL SEARCH REPORT**  
**ON INTERNATIONAL PATENT APPLICATION NO. US 9109067**  
**SA 55197**

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